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Real-time observation of ultrafast Rabi oscillations between excitons and plasmons in J-aggregate/metal hybrid nanostructures

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Surface plasmon polaritons (SPPs), optical excitations at the interface between a metal and a dielectric, carry significant potential for guiding and manipulating light on the nanoscale. Their weak optical nonlinearities, however, hinder active device fabrication, *e.g.*, for all-optical switching or information processing. Recently, strong optical dipole coupling between SPPs and nonlinear quantum emitters with normal mode splittings of up to 700 meV has been demonstrated [1,2]. The predicted ultrafast energy transfer between quantum emitters and SPP fields could be a crucial microscopic mechanism for switching light by light on the nanoscale. Here, we present the first real-time observation of ultrafast Rabi oscillations in a J-aggregate/metal nanostructure, evidencing coherent energy transfer between excitonic quantum emitters and SPP fields. We demonstrate coherent manipulation of the coupling energy by controlling the exciton density on a 10-fs timescale, a step forward towards coherent, all-optical ultrafast plasmonic circuits and devices.